

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent Application of:  
Masahiko Matsukawa et al.

Application No.: 10/743,387

Confirmation No.: 7938

Filed: December 23, 2003

Art Unit: 1742

For: PRETREATMENT METHOD FOR COATING

Examiner: L. L. Zheng

Declaration Under 37 CFR § 1.132

I, Masanobu Futsuhara declare that I am a co-inventor of the inventions disclosed and claimed in the above identified US Patent application and am employed by Nippon Paint Co., Ltd. and I further declare:

1) I have reviewed the Office Action in this application dated March 4, 2010 and the references cited therein.

2) The following experiments were carried out in order to compare the evaluations for corrosion resistance after a cationic electrocoating application using either the chemical conversion coating agent of the present invention or the chemical conversion coating agent described in US Patent 6,203,854 to Affinito. Also, the following experiments were carried out in order to show the significance of limiting the concentration of amino group-containing silane coupling agent to 50-500 ppm as recited in amended claim 29 by comparing the evaluations for corrosion resistance after a cationic electrocoating application using the variables either within the described range or outside the described range. In addition, the following experiments were carried out in order to compare improvements in corrosion resistance obtained by the chemical conversion treatment of the invention of the present application and a zinc phosphate treatment, a widely used chemical conversion treatment technique for iron based materials.

3) The following experiments were conducted by me and/or under my direction and/or supervision.

4) Experiment and evaluation result

<Example 1>

A commercially available cold-rolled steel sheet (SPCC-SD, manufactured by Nippon Testpanel Co., LTD., 70 mm X 150 mm X 0.8 mm) was used as a material, and pretreatment of the coating was applied to the material under the following conditions, followed by application of coating. The material used is an iron based material.

4.1) Pretreatment of coating

Degreasing treatment: The metal material was immersed in 2% by mass of "SURF CLEANER 53" (a degreasing agent manufactured by Nippon Paint Co., Ltd.) at 40°C for 2 minutes.

Rinsing with water after degreasing: The metal material was rinsed for 30 seconds with a spray of running water.

Chemical conversion treatment: A chemical conversion coating agent, having a zirconium concentration of 200 ppm and an amino-group containing silane coupling agent concentration of 200 ppm, was prepared using fluorozirconic acid and KBE-903 (3-aminopropyltriethoxysilane, effective concentration 100%, manufactured by Shin-Etsu Chemical Co., Ltd.) as the amino group-containing silane coupling agent. The pH was adjusted to 4 with sodium hydroxide. The temperature of the chemical conversion coating agent was adjusted to 40°C and the metal material was immersed for 60 seconds.

Rinsing after chemical conversion treatment: The metal material was rinsed for 30 seconds with a spray of running water. Further, the metal material was rinsed for 10 seconds with a spray of ion-exchanged water. Then, electrocoating was applied to the metal material in a wet condition. It should be noted that the amount of the coated material was deemed to be the total amount of metals contained in the chemical conversion coating agent as determined using "XRFI700" (a X-ray fluorescence spectrometer manufactured by Shimadzu Co., Ltd.) after drying the water-rinsed cold-

rolled steel sheet at 80°C for 5 minutes in an electric dryer.

#### 4.2) Coating

Following a treatment of 1 liter of the chemical conversion coating agent per 1 m<sup>2</sup> of the surface of the cold-rolled steel sheet, electrocoating was applied to the surface in such a manner so as to obtain a dried film thickness of 20 μm using "POWERNIX 110" (a cationic electrodeposition coating composition manufactured by Nippon Paint Co., Ltd.) and, after rinsing with water, the metal material was heated and baked at 170°C for 20 minutes and test sheets were prepared.

#### Evaluation Test

[Corrosion resistance characteristics (CCT test; combined cyclic corrosion test)]

The coated surface of the test sheet was scored with a knife in a crosscut pattern to a depth reaching the metal material, and JASO M609-91 combined cyclic corrosion test (a corrosion testing method employed for materials used in automobiles in which one cycle is composed of spraying 5% NaCl solution maintained at 35°C in a salt water spraying chamber kept at 35°C and 95% humidity, drying at 60°C under 20-30% humidity condition for 4 hours, and then incubating at 50°C and under highly humid condition with the humidity of no less than 95% ) was performed for 180 cycles. Subsequently, the formation of rust and blisters from the scored portion of the crosscut pattern was evaluated by observing the maximum width of the rust or blister on both sides of the cut. The Results of evaluation are shown in table 1.

A: The maximum width of the rust or blister formed is no more than 10mm on both sides of the cut.

B: The maximum width of the rust or blister formed is no less than 10mm but no more than 12mm on both sides of the cut.

C: The maximum width of the rust or blister formed is no less than 12mm but no more than 18mm on both sides of the cut.

D: The maximum width of the rust or blister formed is no less than 18mm on both sides of the cut.

<Example 2, Comparative Examples 1-5>

Test sheets were prepared in the same manner as in Example 1 except for changing the concentration of zirconium and the solid portion of amino group-containing silane coupling agent to those shown in Table 1. The sheets were then evaluated. In the specification of the present application, the total amount of metals contained in the chemical conversion coating is described as the coat amount (paragraph 0067 of the specification of the present application). Here, for reference, the amount of Si in the coat is also shown, reflecting the presence of aminosilane that is incorporated into the chemical conversion coat.

<Referring Example>

Degreasing treatment and subsequent water rinsing of the material was carried out in the same manner as in Example 1, followed by surface adjustment at ambient temperature for 30 seconds using SURFFINE 5N-8M (manufactured by Nippon Paint Co., Ltd.). Chemical conversion treatment was then performed by immersion treatment for 2 minutes at 35°C using SURFDINE SD-6350 (manufactured by Nippon Paint Co., Ltd.), and coating was done using the same method as in Example 1.

Table 1: Comparison of the experimental data

|                       | zirconium concentration (ppm)                | concentration amino group-containing silane coupling agent (ppm) | coat amount (mg/m <sup>2</sup> ) | amount of Si contained in the coat (mg/m <sup>2</sup> ) | corrosion resistance characteristics (CCT) (mm) |
|-----------------------|--|--|----------------------------------|---|---|
| Example 1             | 200  | 200  | 35                               | 3   | A   |
| Example 2             | 500  | 500  | 40                               | 3   | A   |
| Comparative Example 1 | 200  | 800  | 20                               | 5   | C   |
| Comparative Example 2 | 10   | 200  | 10                               | 4   | C   |
| Comparative Example 3 | 200  | 10   | 60                               | 0.3   | C   |
| Comparative Example 4 | 2000   | 3000   | 5                                | 5   | D   |
| Comparative Example 5 | 500  | 1000   | 15                               | 4   | C   |
| Referring Example     | zinc phosphate chemical conversion treatment |  |                                  |   | B   |

### 5) Discussion

As can be clearly seen in Table 1, Examples 1 and 2 that use the chemical conversion coating agent of the present invention exhibited favorable corrosion resistance characteristics compared with the Comparative Examples 1-5 that use various chemical conversion coating agents not conforming to the requirements of the present invention. Notably, Comparative Example 4, using the chemical conversion coating agent having the preferred concentration of zirconium and amino group-containing silane coupling agent of the invention described in Affinito, exhibited a lack of adequate corrosion resistance characteristics, suggesting that the preferred chemical conversion coating agent of the invention described in Affinito is not able to provide a favorable chemical conversion treatment for materials based on iron. In contrast, the data in Table 1 above demonstrate that the use of chemical conversion coating agent having amino group-containing silane coupling agent at a concentration of 50-500 ppm and zirconium at a concentration of 50-2000 ppm (in other words, the chemical conversion coating agent of the invention of the present application), provides for favorable chemical conversion treatment on iron based materials.

All statements made herein of my own knowledge are true. All statements made on information and belief are believed to be true. These statements were made with the knowledge that willful false statements and the like so made are punishable by fine, imprisonment, or both, under 18 U.S.C. 1001 and may jeopardize the validity of the application or any patent issuing thereon.

Respectfully submitted,

Date:

Masanobu Futsuhara

27<sup>th</sup>/June/2011

Futsuhara Masanobu